Collective two-photon absorption of a squeezed electromagnetic field

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We report a theoretical study of two-quantum interaction processes of a system of atoms and a squeezed electromagnetic field. We consider the way absorption processes depend on the quantum fluctuations of the squeezed light and on the method of (amplitude or phase) squeezing. We show that the rate of absorption of the applied electromagnetic field is higher in the case of phase squeezing than for amplitude squeezing and that the two-quantum Rabi frequency takes on its largest value in the case of phase squeezing and in the case of amplitude squeezing its lowest. We show that if the fluctuations of the electromagnetic field strength are of the order of the average electromagnetic field strength the quantum fluctuations are in the case of amplitude squeezing able to stop the two-quantum nutation of the system.

1. INTRODUCTION

A number of experimental studies devoted to the generation of incoherent kinds of electromagnetic fields (EMF) have been carried out in the last decade; among them some paid special attention to fields with reduced fluctuations of one of the phase quadratures of the electromagnetic field strength:¹⁻³

$$X_1 = a_k^+ + a_k, \quad X_2 = i(a_k^+ - a_k),$$

where a_k^+ (a_k) are the EMF creation (annihilation) operators satisfying the commutation rules

$$[a_{\mathbf{k}}, a_{\mathbf{k}'}^+] = \delta_{\mathbf{k}\mathbf{k}'}, \quad [a_{\mathbf{k}}, a_{\mathbf{k}'}] = 0.$$

Such states have been called squeezed states of the electromagnetic field. One possible definition of a squeezed EMF is the following:⁴

 $|s\rangle_{\mathbf{k}} = T_{\mathbf{k}}(\alpha) D_{\mathbf{k}}(\xi) |0\rangle_{\mathbf{k}},$

where $|0\rangle_{\mathbf{k}}$ is the vacuum state of the **k** mode of the EMF,

$$T_{\mathbf{k}}(\alpha) = \exp(\alpha a_{\mathbf{k}}^{+} - \alpha^{*} a_{\mathbf{k}}),$$

$$D_{\mathbf{k}}(\xi) = \exp(\frac{1}{2}\xi^{*} a_{\mathbf{k}}^{2} - \frac{1}{2}\xi a_{\mathbf{k}}^{+2})$$

are the shift and squeeze operators in the X_1X_2 phase plane, and

$$\alpha = |\alpha| \exp(i\varphi_{\alpha}), \quad \xi = r \exp(i\varphi_{r})$$

are complex parameters, $0 \le r$, $|\alpha| < \infty$, $0 \le \varphi_{\alpha}, \varphi_r < 2\pi$. The EMF operators transform as follows under the action of the shift and squeeze operators:⁴

$$T_{\mathbf{k}'}^{-1}(\alpha)a_{\mathbf{k}}^{+}T_{\mathbf{k}'}(\alpha) = a_{\mathbf{k}}^{+} + \alpha^{*}\delta_{\mathbf{k}\mathbf{k}'}, \qquad (1a)$$
$$D_{\mathbf{k}'}^{-1}(\xi)a_{\mathbf{k}}^{+}D_{\mathbf{k}'}(\xi) \qquad \qquad (1a)$$

$$=a_{\mathbf{k}} + [1 + \delta_{\mathbf{k}\mathbf{k}'}(\operatorname{ch} r - 1)] - \delta_{\mathbf{k}\mathbf{k}'}a_{\mathbf{k}}\operatorname{sh} r \exp(-i\varphi_{r}). \quad (1b)$$

A squeezed EMF can be considered as a field consisting of a classical component and a fluctuating component also called the squeezed vacuum.

After squeezed EMF states had been detected experimentally, people became interested in studying interaction processes between squeezed light and matter (atoms, molecules). For instance, in Refs. 5 to 7 the behavior of a system of atoms reaching single-photon resonance with a squeezed EMF was studied qualitatively. Since two-photon processes depend strongly on the quantum fluctuations of the applied EMF⁸ the study of two-quantum absorption of squeezed light attracted special interest because it is well known that the statistical properties of squeezed EMF can vary to a large extent with changes in the parameters α and ξ .⁴ For instance, it was shown in Ref. 9 that the probability for twoquantum absorption of squeezed light depends on the degree of the squeezing and how it is done (amplitude or phase squeezing). In the case of amplitude squeezing the rate of two-photon absorption decreases with increasing intensity of the coherent component of the squeezed EMF. Using the model interaction Hamiltonian

$$H_{int}' = -\vec{q} : \mathbf{E}(t)\mathbf{E}(t)$$
(2)

[here \ddot{q} is the two-photon tensor operator of the atomic subsystem and E(t) the electric field strength] the governing equation for the statistical operator of a single atom in the field of a wide-band squeezed light beam was found in Ref. 10. It was assumed there that the classical component of the squeezed EMF strength was much larger than the fluctuating component. We note also that the approach using the model Hamiltonian (2) assumes a weak applied EMF strength since in a strong EMF the atoms are able to relax to the ground state not only through spontaneous decay and reemission of two photons of the external EMF, but also through scattering of the applied EMF and emission of anti-Stokes photons.¹¹

In the present paper we consider the collective two-photon interaction process of a concentrated system of atoms and a squeezed EMF. In contrast to Ref. 10 we use the Hamiltonian of a three-level system with dipole-forbidden transitions between the first two energy levels.¹¹ The advantage of such an approach was discussed in Ref. 12. Indeed, the method used makes it possible to take the two-quantum processes in the system fully into account, including scattering processes and not to be restricted to a weak EMF strength while also, in contrast to Ref. 10, it is possible to consider any magnitude of fluctuations in the EMF strength. In particular, we consider the case when the amplitude of the quantum fluctuations of the EMF strength is of the same order of magnitude as the average value of the EMF strength. In that situation the frequency of the two-photon nutation depends strongly on the way the EMF is squeezed; we show that in

the case of amplitude squeezing the quantum fluctuations of the EMF are able to halt the two-photon nutation of the system. We obtain in the present paper the explicit form of the "loss" coefficient describing the change in the number of photons in a beam of squeezed light as the result of the interaction with an atomic system.

We consider several special cases including the twophoton interaction between a system of atoms and a coherent EMF and we show that in that case the possibility for a phase transition of the atomic system depends on the number of atoms.

The content of the paper is arranged as follows. In the second section we transform the Hamiltonian of a three-level system¹¹ into an effective Hamiltonian which enables us to use a lemma on the elimination of the boson operators of the EMF;¹¹ using this we obtain in the third section the governing equation for the statistical operator of the atomic subsystem. In the fourth section we consider the change in the number of photons in a beam of squeezed light as the result of the absorption of light by the atomic system for which we introduce a loss coefficient expressed in terms of the correlator of the atomic subsystem whose stationary values we find by solving the governing equation for the statistical operator of the system of atoms.

2. DERIVATION OF THE EFFECTIVE HAMILTONIAN OF THE SYSTEM

We consider a system of N atoms in two-photon resonance with a squeezed EMF. For simplicity we assume that the linear dimensions of the system are much smaller than the wavelength of the radiation. The Hamiltonian has the form¹¹

$$H = H_{A} + H_{ph} + H_{ini}, \qquad H_{A} = \sum_{\beta=1}^{3} \varepsilon_{\beta} U_{\beta}^{\beta},$$

$$H_{ph} = \sum_{k} \hbar \omega_{k} a_{k}^{+} a_{k},$$

$$H_{ini} = i \sum_{k} \sum_{\beta=1}^{2} g_{k} d_{3\beta} (U_{\beta}^{3} + U_{3}^{\beta}) (a_{k}^{+} - a_{k}).$$
(3)

Here $\omega_{\mathbf{k}}$ is the frequency of the EMF, ε_{β} is the energy of the $|\beta\rangle$ level, and $\mathbf{d}_{\gamma\beta}$ and U^{γ}_{β} are, respectively, the dipole moment matrix element and the collective operator for a transition between the $|\gamma\rangle$ and the $|\beta\rangle$ levels. The U^{γ}_{β} operators satisfy the commutation relation

$$\begin{split} [U_{\beta}^{\tau}, U_{\beta'}^{\tau'}] = & U_{\beta'}^{\tau} \delta_{\tau'\beta} - U_{\beta}^{\tau'} \delta_{\tau\beta'}, \\ \mathbf{g}_{\mathbf{k}} = (2\pi \hbar \omega_{\mathbf{k}}/V)^{\eta_2} \mathbf{e}_{\lambda}, \end{split}$$

V is the quantization volume, and $\omega_{\mathbf{k}}$ and \mathbf{e}_{λ} ($\lambda = 1,2$) are the frequency and polarization of a photon with wavevector **k**.

Let us consider a Schrödinger operator Q^{Sch} . Its average can be written in the form

$$\langle Q \rangle = \operatorname{Sp} \{ \rho(t) Q^{\operatorname{Sch}} \},$$

$$\rho(t) = S(t) \rho_A(0) \otimes \rho_{ph}(0) S^+(t),$$

where $\rho(t)$ is the statistical operator of the "atom + field" system, $\rho_A(0)$ is the statistical operator of the atomic subsystem at time t = 0,

$$\rho_{ph}(0) = \prod_{\mathbf{k}} T_{\mathbf{k}_{0}}(\alpha) D_{\mathbf{k}_{0}}(\xi) |0\rangle_{\mathbf{k}\mathbf{k}} \langle 0| D_{\mathbf{k}_{0}}^{-1}(\xi) T_{\mathbf{k}_{0}}^{-1}(\alpha)$$

the statistical operator of the photon subsystem, and

$$S(t) = \exp(-iHt/\hbar)$$

the evolution operator.

Using the commutativity of the operator $\rho_A(0)$ with the $T_{\mathbf{k}_0}(\alpha)$ and $D_{\mathbf{k}_0}(\xi)$ operators we can, after cyclic permutations under the trace sign, write

$$\langle Q \rangle = \operatorname{Sp} \{ \rho_A(0) \otimes W_{ph}(0) Q(t) \},$$
(4)

$$W_{ph}(0) = \prod_{\mathbf{k}} |0\rangle_{\mathbf{k} \mathbf{k}} \langle 0|, \qquad (5)$$

$$Q(t) = D_{\mathbf{k}_0}^{-1}(\xi) T_{\mathbf{k}_0}^{-1}(\alpha) \tilde{S}^+(t) Q^{sh} S(t) T_{\mathbf{k}_0}(\alpha) D_{\mathbf{k}_0}(\xi).$$
(6)

Equation (6) is a new representation of the Q operator in which the action of the shift and squeezing operators is transferred to the evolution operator S(t): the latter can be written in the form

$$S(t) = S_0(t)\tilde{S}(t),$$

$$S_0(t) = \exp\left(-i\omega_0\sum_{\mathbf{k}}a_{\mathbf{k}^{\top}}a_{\mathbf{k}}\right),$$

$$\tilde{S}(t) = T\exp\left[-\frac{i}{\hbar}\int_0^t d\tau S_0^+(\tau)H(\tau)S_0(\tau)\right],$$

where ω_0 is the frequency of the external EMF and T the chronological ordering.

Differentiating Eq. (6) using the relations

$$\frac{\partial \widetilde{S}(t)}{\partial t} = -\frac{i}{\hbar} S_0^+(t) H(t) S_0(t) \widetilde{S}(t),$$

 $\hat{1} = T_{\mathbf{k}_0}(\alpha) D_{\mathbf{k}_0}(\xi) D_{\mathbf{k}_0}(\xi) T_{\mathbf{k}_0}(\alpha)$

(here $\hat{1}$ is the unit operator), we get the following equation:

$$\frac{dQ(t)}{dt} = \frac{\partial Q(t)}{\partial t} + \frac{i}{\hbar} [H^{eff}, Q(t)], \qquad (7)$$

$$H^{eff} = H_A + \sum_{\mathbf{k}} \hbar (\omega_{\mathbf{k}} - \omega_0) a_{\mathbf{k}}^+ (t) a_{\mathbf{k}} (t)$$

$$+ i \sum_{\mathbf{k}} \sum_{\mathbf{k}}^2 \mathbf{g}_{\mathbf{k}} \mathbf{d}_{\mathbf{3}\beta} [U^{3}_{\beta} (t) + U^{3}_{\beta} (t)]$$

$$\frac{1}{k} \int_{\beta=1}^{\infty} \chi(\mathbf{k}, t) a_{\mathbf{k}}^{+}(t) - \chi^{*}(\mathbf{k}, t) a_{\mathbf{k}}(t) + \delta_{\mathbf{k}\mathbf{k}_{0}}[\alpha^{*}(t) - \alpha(t)] \}, \quad (8)$$

$$\chi(\mathbf{k}, t) = 1 + \delta_{\mathbf{k}\mathbf{k}_{0}} \{\operatorname{ch} r - 1 + \operatorname{sh} r \exp\left[-i(2\omega_{0}t - \varphi_{r})\right] \}, \quad \alpha(t) = |\alpha| \exp\left[-i(\omega_{0}t - \varphi_{\alpha})\right].$$

The representation (6) is convenient in that the statistical operator of the photon subsystem has in that representation the form (5) which makes it possible to use the method of elimination of the boson operators of the \mathbf{EMF}^{11} when deriving the governing equation for the statistical operator of the atomic subsystem.

3. GOVERNING EQUATION FOR THE STATISTICAL OPERATOR OF THE ATOMIC SUBSYSTEM

We average Eq. (7) over the whole of the (atom + field) system and we shall assume Q(t) to be a Hermitean operator of the atomic subsystem; using (8) we get as a re-

$$\left\langle \frac{dQ(t)}{dt} \right\rangle + \frac{i}{\hbar} \left\langle [Q(t), H_A] \right\rangle = \mathcal{D}_1(t) + \mathcal{D}_2(t) + \text{h.c.}, \qquad (9)$$

where

$$\mathcal{D}_{1}(t) = \alpha^{\star}(t) \sum_{\beta=1}^{2} \frac{\mathbf{g}_{\mathbf{k}_{0}} \mathbf{d}_{3\beta}}{\hbar} \langle [Q(t), U_{3}^{\beta}(t) + U_{\beta}^{3}(t)] \rangle,$$

$$\mathscr{D}_{2}(t) = \sum_{\mathbf{k}} \sum_{\beta=1}^{\mathbf{g}_{\mathbf{k}} \mathbf{d}_{3\beta}} \chi(\mathbf{k}, t) \langle a_{\mathbf{k}}^{+}(t) [Q(t), U_{3}^{\beta}(t) + U_{\beta}^{3}(t)] \rangle.$$

In deriving this equation we use the expansion of the right-hand side of Eq. (9) in a perturbation theory series with small parameter¹³

$$\varepsilon(\mathbf{k}) = \mathbf{g}_{\mathbf{k}} \mathbf{d}_{\mathbf{3}\mathbf{\beta}} / \hbar \left(\omega_{\mathbf{3}\mathbf{\beta}} \pm \omega_{\mathbf{k}} \right), \tag{10}$$

where $\omega_{\gamma\beta}$ is the frequency of the transition between the $|\gamma\rangle$ and $|\beta\rangle$ levels. We assume that the conditions

$$\epsilon(\mathbf{k}_0) |\alpha| \leq 1, \quad \epsilon(\mathbf{k}_0) \operatorname{sh} r \leq 1$$

are always satisfied. First of all we find the first-order terms of the perturbation theory. We consider the term $\mathscr{D}_1(t)$. Since the $|3\rangle$ level is unoccupied at the moment the interaction between the atomic and the photon subsystems is switched on we have $U_3^3(0) = 0$ and the applied EMF is not in resonance with respect to the transitions between the $|3\rangle$ and $|\beta\rangle$ ($\beta = 1,2$) levels and the $|3\rangle$ state is a virtual one. It is thus necessary to eliminate the transitions connected with the $|3\rangle$ level. To do this we formulate the equation of motion for the $U_3^\beta(t)$ operator and write down its formal solution:

$$U_{3}^{\beta}(t) = U_{3}^{\beta}(0) \exp\left(-i\omega_{3\beta}t\right) - \sum_{\mathbf{k}} \sum_{\tau=1}^{2} \frac{\mathbf{g}_{\mathbf{k}} \mathbf{d}_{3\tau}}{\hbar} \int_{0}^{t} d\tau \left[U_{3}^{3}(t-\tau)\delta_{\beta\tau}\right] + U_{\tau}^{\beta}(t-\tau) \exp\left(-i\omega_{3\beta}\tau\right) \left\{\chi(\mathbf{k},t-\tau)a_{\mathbf{k}}^{+}(t-\tau)\right. -\chi^{*}(\mathbf{k},t-\tau)a_{\mathbf{k}}(t-\tau) + \delta_{\mathbf{k}\mathbf{k}_{0}}\left[\alpha^{*}(t-\tau)-\alpha(t-\tau)\right]\right\}.$$
(11)

We substitute (11) into the expression for $\mathscr{D}_1(t)$; one notes easily that in first order of perturbation theory one needs only take into account the integral term in (11) which contains $\alpha^*(t-\tau)$ and $\alpha(t-\tau)$. Evaluating the integral in the Born approximation and dropping fast oscillating terms, we find the first-order correction:

$$\mathcal{D}_{1}^{(1)}(t) = i\Omega(\alpha) \langle [U_{2}^{1}(t), Q(t)] \rangle \exp(2i\omega_{0}t), \qquad (12)$$

where

$$\Omega(\alpha) = |\alpha|^{2} \frac{(\mathbf{g}_{\mathbf{k}_{0}}, \mathbf{d}_{31})(\mathbf{g}_{\mathbf{k}_{0}}, \mathbf{d}_{32})}{2\hbar^{2}} \theta^{(-)}(\omega_{\mathbf{k}_{0}}) \exp(-2i\varphi_{\alpha}), \quad (13)$$
$$\theta^{(\pm)}(x) = \frac{\omega_{31} + \omega_{32}}{(\omega_{31} \pm x)(\omega_{32} \mp x)}.$$

Expression (12) describes the two-quantum nutation of the system with a frequency $\Omega(\alpha)$,¹¹ caused by the coherent component of the squeezed EMF.

The expression for $\mathcal{D}_2(t)$ contains the EMF operator $a_{\mathbf{k}}^+(t)$ which must be expressed in terms of the atomic subsystem operators. To do this we write down the equation of motion for the $a_{\mathbf{k}}^+(t)$ operator:

$$a_{k}^{+}(t) = a_{k}^{e+}(t) + a_{k}^{s+}(t), \qquad (14a)$$

$$a_{k}^{e+}(t) = a_{k}^{e+}(0) \exp(i\omega_{k}t), \qquad (14b)$$

$$a_{\mathbf{k}^{s+}}(t) = \sum_{\beta=1}^{2} \frac{g_{\mathbf{k}} \mathbf{d}_{s\beta}}{\hbar} \int_{0}^{t} d\tau \chi(\mathbf{k}, t-\tau) \exp(i\omega_{\mathbf{k}}\tau) \\ \times [U_{s}^{\beta}(t-\tau) + U_{\beta}^{3}(t-\tau)].$$
(14c)

We substitute (14) into the expression for $\mathscr{D}_2(t)$; using the form (5) of the statistical operator of the photon subsystem and also the property $\langle 0|a_{\mathbf{k}}^{e^+} = 0$ we find

$$\mathcal{D}_{2}(t) = \sum_{\mathbf{k}} \sum_{\beta_{1}=1}^{2} \sum_{\beta_{2}=1}^{2} \frac{(\mathbf{g}_{\mathbf{k}} \mathbf{d}_{3\beta_{1}}) (\mathbf{g}_{\mathbf{k}} \mathbf{d}_{3\beta_{2}})}{\hbar^{2}} \chi(\mathbf{k}, t) \int_{0}^{1} d\tau \chi(\mathbf{k}, t-\tau) \\ \times \exp(i\omega_{\mathbf{k}}\tau) \langle [U_{3}^{\beta_{2}}(t-\tau) + U_{\beta_{2}}^{3}(t-\tau)] \\ \times [Q(t), U_{3}^{\beta_{1}}(t) + U_{\beta_{1}}^{3}(t)] \rangle.$$
(15)

If we use the Born approximation to evaluate the integral on the right-hand side of (15) we get the first-order term of the expansion of $\mathscr{D}_2(t)$ in a perturbation theory series with small parameter (10):

$$\mathcal{D}_{2}^{(1)}(t) = -i\Omega(r) \langle [U_{2}^{(1)}(t), Q(t)] \rangle \exp((2i\omega_{0}t)), \qquad (16)$$

$$\Omega(r) = \operatorname{sh} r \operatorname{ch} r \frac{(\mathbf{g}_{\mathbf{k}_{0}}\mathbf{d}_{31})(\mathbf{g}_{\mathbf{k}_{0}}\mathbf{d}_{32})}{2\hbar^{2}} \theta^{(-)}(\omega_{\mathbf{k}_{0}}) \exp(-i\varphi_{r}). \quad (17)$$

If we then use (12) and (16), Eq. (9) takes in first order of perturbation theory the form

$$\left\langle \frac{dQ^{(1)}(t)}{dt} \right\rangle + \frac{i}{\hbar} \langle [Q(t), H_A] \rangle$$

= $i\Omega(r, \alpha) \langle [U_2^{+}(t), Q(t)] \rangle \exp(2i\omega_0 t) + \text{h.c.},$ (18)

$$\Omega(r, \alpha) = \Omega(\alpha) - \Omega(r).$$
(19)

It follows from Eqs. (18) and (19) that in the case of a two-photon interaction with the squeezed EMF the Rabi frequency $\Omega(r,\alpha)$ depends on the squeezing parameter r. The nutation frequency $\Omega(r,\alpha)$ then depends strongly on the quantum fluctuations of the applied EMF. For instance, in the case of the strongest photon bunching, which is reached for a squeezing phase $2\varphi_{\alpha} = \varphi_r + \pi$, the Rabi frequency $\Omega(r,\alpha)$ takes on its largest possible value for given r and α . When the photon bunching in the squeezed EMF weakens the nutation frequency $\Omega(r,\alpha)$ decreases and reaches its smallest value in the case of amplitude squeezing, $2\varphi_{\alpha} = \varphi_r$, when the photons are least bunched, as is well known⁴ (antibunching is also possible, if $|\alpha|^2 > (\sinh^3 r + \sinh r \cosh r)^2 / 2(\cosh r - \sinh r)$). In that case the interaction of a system of atoms with a squeezed EMF in which the amplitude of the quantum fluctuations of the EMF strength is of the order of the average value of the EMF strength or, more precisely, when $\sinh r \cosh r = |\alpha|^2$, is of special interest; in that case the quantum fluctuations of the applied EMF stop the nutation in the system $(\Omega(r,\alpha)=0).$

Since we have not restricted the external EMF to large intensities we must take into account not only the nutation processes, but also the damping processes in the system, in particular, the spontaneous decay of the $|2\rangle$ level. To do this we must consider second-order terms of the perturbation theory. For instance, in order to obtain the second-order correction to $\mathcal{D}_1(t)$ we must take into account the integral terms containing $a_k^+(t-\tau)$ and $a_k(t-\tau)$ in expression (11) which was earlier substituted into $\mathcal{D}_1(t)$. Using Eqs. (14) and also the expression

$$U_{3}^{\beta}(t-\tau) = U_{3}^{\beta}(t) \exp(i\omega_{3\beta}\tau)$$

$$+ \sum_{\mathbf{k}} \sum_{\tau=1}^{2} \frac{g_{\mathbf{k}} d_{3\tau}}{\hbar} \int_{0}^{\tau} d\mu [U_{3}^{3}(t-\tau+\mu)\delta_{\beta\tau}]$$

$$+ U_{\tau}^{\beta}(t-\tau+\mu) \exp(i\omega_{3\beta}\mu)] \{\chi(\mathbf{k}, (t-\tau+\mu)a_{\mathbf{k}}^{+}(t-\tau+\mu) - -\chi^{*}(\mathbf{k}, t-\tau+\mu)a_{\mathbf{k}}(t-\tau+\mu) + \delta_{\mathbf{k}\mathbf{k}} [\alpha^{*}(t-\tau+\mu)-\alpha(t-\tau+\mu)]\},$$

(20) obtained as the result of solving the equation of motion for the $U_3^{\beta}(t-\tau)$ operator, and also using the lemma about the elimination of boson operators from Ref. 11, we find the second-order corrections to $\mathscr{D}_1(t)$ and $\mathscr{D}_2(t)$. After some

transformations in the final form we get the equation

$$\left\langle \frac{d\hat{Q}}{dt} \right\rangle + i\delta \langle [\hat{Q}, R_z] \rangle$$

$$= i\Omega(r, \alpha) \langle [R^-, \hat{Q}] \rangle + a(r, \alpha) \langle R^+[\hat{Q}, R^-] \rangle$$

$$- b(r, \alpha) \langle [\hat{Q}, R^-] R^+ \rangle - d(r, \alpha) \langle R^+[\hat{Q}, R^+] \rangle$$

$$- d^*(r, \alpha) \langle R^-[\hat{Q}, R^-] \rangle + \text{h.c.},$$

$$(21)$$

where the quantity $\delta = \omega_{21} - 2\omega_0$ determines the frequency mismatch of the resonance while R^{\pm} and R_z are quasispin operators connected with the earlier atomic subsystem operators through the relations

$$R^{-} = U(t)U_{2^{1}}(t)U^{+}(t), \quad R^{+} = (R^{-})^{+}, \quad 2R_{z} = [R^{+}, R^{-}],$$

$$\bar{Q} = U(t)Q(t)U^{+}(t),$$

$$U(t) = \exp \{i\omega_{0}[U_{2^{2}}(t) - U_{1^{1}}(t)]t\}.$$

The coefficients of Eq. (21) have the form

$$a(r, \alpha) = \sum_{\mathbf{k}_{1}} \sum_{\mathbf{k}_{2}} \frac{(\mathbf{g}_{\mathbf{k}_{1}} \mathbf{d}_{31})^{2} (\mathbf{g}_{\mathbf{k}_{2}} \mathbf{d}_{32})^{2}}{2\hbar^{i}} \\ \times \{\theta^{(-)^{2}} (\omega_{\mathbf{k}_{2}}) [1 + 2\delta_{\mathbf{k}_{0}\mathbf{k}_{1}} (|\alpha|^{2} + \mathrm{sh}^{2} r) \\ + \delta_{\mathbf{k}_{0}\mathbf{k}_{1}} \delta_{\mathbf{k}_{0}\mathbf{k}_{2}} \mathrm{sh}^{2} r (2|\alpha|^{2} + \mathrm{sh}^{2} r)]\varphi(\omega_{\mathbf{k}_{1}} + \omega_{\mathbf{k}_{2}} - \omega_{21}) \\ + 2\theta^{(+)^{2}} (\omega_{0}) \delta_{\mathbf{k}_{2}\mathbf{k}_{0}} \\ \times [|\alpha|^{2} + \mathrm{sh}^{2} r] \varphi(\omega_{\mathbf{k}_{1}} - \omega_{\mathbf{k}_{0}} - \omega_{21}) \}, \qquad (22a)$$

$$b(r,\alpha) = \sum_{\mathbf{k}_{1}} \sum_{\mathbf{k}_{2}} \delta_{\mathbf{k}_{1}\mathbf{k}_{0}} \delta_{\mathbf{k}_{2}\mathbf{k}_{0}} \frac{(\mathbf{g}_{\mathbf{k}_{1}}\mathbf{d}_{\mathbf{3}1})^{2} (\mathbf{g}_{\mathbf{k}_{2}}\mathbf{d}_{\mathbf{3}2})^{2}}{2\hbar^{4}} \theta^{(-)2}(\omega_{\mathbf{k}_{2}}) \mathrm{sh}^{2} r.$$

$$\times [2|\alpha|^{2} + \operatorname{sh}^{2} r] \varphi(\omega_{\mathbf{k}_{1}} + \omega_{\mathbf{k}_{2}} - \omega_{21}), \qquad (22b)$$

$$\sum \sum \sum (\mathbf{g}_{\mathbf{k}_{1}} \mathbf{d}_{31})^{2} (\mathbf{g}_{\mathbf{k}_{2}} \mathbf{d}_{32})^{2} \rho(-)^{2} \langle \mathbf{g}_{\mathbf{k}_{2}} \mathbf{d}_{32} \rangle^{2} \rho(-)^{2} \rho(-)^{2}$$

$$d(r, \alpha) = \sum_{\mathbf{k}_1} \sum_{\mathbf{k}_2} \delta_{\mathbf{k}_1 \mathbf{k}_0} \delta_{\mathbf{k}_2 \mathbf{k}_0} \frac{(\mathbf{g}_{\mathbf{k}_1} \mathbf{u}_{31})^{-} (\mathbf{g}_{\mathbf{k}_2} \mathbf{u}_{32})}{2\hbar^4} \theta^{(-)^2}(\omega_{\mathbf{k}_2}) \operatorname{sh} r \operatorname{ch} r$$

 $\times [2|\alpha|^2 \exp(2i\varphi_{\alpha}) - \operatorname{sh} r \operatorname{ch} r \exp(i\varphi_{r})] \varphi(\omega_{\mathbf{k}_{1}} + \omega_{\mathbf{k}_{2}} - \omega_{21}).$ (22c)

The function

$$\varphi(x) = \frac{\Gamma/2}{x^2 + (\Gamma/2)^2}$$

has a Lorentzian form where Γ is the spectral width of the $|2\rangle$ level.

Consistently taking the first- and second-order terms of the expansion series of the right-hand side of Eq. (9) into account we thus obtained a kinetic equation for the atomic subsystem operators. Since Eq. (21) does not contain the EMF operators the averaging is only over the atomic subsystem states. Transferring the temporal evolution to the statistical operator of the atomic subsystem we easily find the following governing equation:

$$\frac{d\rho_A}{dt} + i\delta[R_z, \rho_A] = i\Omega(r, \alpha) [\rho_A, R^-] + a(r, \alpha) [R^-, \rho_A R^+]$$
$$-b(r, \alpha) [R^-, R^+\rho_A] - d(r, \alpha) [R^+, \rho_A R^+]$$
$$-d^{\bullet}(r, \alpha) [R^-, \rho_A R^-] + \text{h.c.}$$
(23)

Let us consider in det: I the second-order processes described by the $a(r,\alpha)$ and $b(r,\alpha)$ coefficients. It follows from Eqs. (22) that in each relaxation and excitation act of the system the following processes are possible: the emission of two spontaneous photons (Fig. 1,1), the scattering of the applied EMF at the frequencies $\omega_k = \omega_{21} \pm \omega_0$ (Figs. 1,2-1,5), and also induced transitions involving two quanta of the squeezed vacuum (Figs. 1,6; 1,8), or involving one coherent photon and one photon from the squeezed vacuum (Figs. 1,7; 1,9).

We note that although the form of Eq. (23) does not differ from the one obtained in Ref. 10 there are a number of important differences. The equation from Ref. (10) does not take into account the processes involving the scattering of the external EMF at the frequencies $\omega_{\mathbf{k}} = \omega_{21} \pm \omega_0$ (Figs. 1,2; 1,5), or the processes involving one coherent photon and one photon from the squeezed vacuum (Figs. 1,7; 1,9). It is therefore impossible in the framework of Ref. 10 to obtain in the limit as $r \rightarrow 0$ the equation¹¹

$$\frac{d\rho_A}{dt} + i\delta[R_2, \rho_A] = i\Omega(\omega)[\rho_A, R^-] + a(0, \alpha)[R^-, \rho_A R^+] + \text{h.c.},$$
(24)

describing the two-quantum interaction of a system of atoms with the coherent EMF mode. Moreover, the equation in Ref. 10 is obtained under the condition $|\alpha|^2 \gg \sinh r \cosh r$ and hence one cannot consider the case of amplitude squeezing of the applied EMF when $|\alpha|^2 = \sinh r \cosh r$ and the nutation frequency becomes zero. Equation (23) takes in that case the form

$$\frac{d\rho_{A}}{dt} + i\delta[R_{z},\rho_{A}]$$

$$= a(r,\alpha)[R^{-},\rho_{A}R^{+}] - b(r,\alpha)[R^{-},R^{+}\rho_{A}] - d(r,\alpha)$$

$$\times [R^{+},\rho_{A}R^{+}] - d^{*}(r,\alpha)[R^{-},\rho_{A}R^{-}] + \text{h.c.}$$
(25)

The two-quantum absorption processes of squeezed light depend strongly on the ratio of the magnitude of the



FIG. 1. The thick straight lines indicate the photons of the classical component, the straight lines the photons of the fluctuating component of the squeezed EMF, and the wavy lines the spontaneously created photons.

spectral width Γ of the $|2\rangle$ level and the spectral width K of the applied EMF beam.9 The necessity to take into account the spectral widths Γ and K also follows from the form of the coefficients (22), part of which contains under the summation sign over \mathbf{k}_1 and \mathbf{k}_2 two Kronecker delta symbols and the φ function which for small Γ behaves like a Dirac delta function. It is thus advisable to consider two limiting cases: large spectral width of the excited level of the atomic system, $\Gamma \gg K$, and small spectral width, $\Gamma \ll K$. Changing in (22) from a summation to an integration we get after some simple transformations

$$a(r,\alpha) = a_1 + a_2(r,\alpha) + a_3(r,\alpha) + a_4(r,\alpha),$$
 (26)

$$a_{1} = \frac{2d_{31}^{2}d_{32}^{2}}{9\pi c^{6}\hbar^{2}} \int_{0}^{\omega_{21}} dx \theta^{(-)2}(x) x^{3}(\omega_{21}-x)^{3}, \qquad (26a)$$

$$a_{z}(r,\alpha) = \frac{2(\mathbf{g}_{\mathbf{k}_{0}}\mathbf{d}_{31})(\mathbf{g}_{\mathbf{k}_{0}}\mathbf{d}_{32})d_{31}d_{32}}{3c^{3}\hbar^{3}} \times \theta^{(-)2}(\boldsymbol{\omega}_{\mathbf{k}_{0}})(\boldsymbol{\omega}_{21}-\boldsymbol{\omega}_{0})^{3}[|\alpha|^{2}+\mathrm{sh}^{2}r], \qquad (26b)$$

$$a_{3}(r,\alpha) = \frac{2(\mathbf{g}_{k_{0}}\mathbf{d}_{31})(\mathbf{g}_{k_{0}}\mathbf{d}_{32})d_{31}d_{32}}{3c^{3}\hbar^{3}}$$

$$\times \theta^{(+)^2}(\omega_{\mathbf{k}_0}) (\omega_{21} + \omega_0)^3 [|\alpha|^2 + \mathrm{sh}^2 r], \qquad (26c)$$

$$a_{4}(r,\alpha) = \frac{(\mathbf{g}_{\mathbf{k}_{0}}\mathbf{d}_{31})^{2}(\mathbf{g}_{\mathbf{k}_{0}}\mathbf{d}_{32})^{2}}{2\hbar^{4}} \times \theta^{(-)2}(\omega_{\mathbf{k}_{0}})\operatorname{sh}^{2}r[2|\alpha|^{2} + \operatorname{sh}^{2}r]L(\delta), \quad (26d)$$

$$b(r,\alpha) = a_4(r,\alpha), \qquad (27)$$

$$d(r, \alpha) = \frac{(\mathbf{g}_{\mathbf{k}_0} \mathbf{d}_{31})^2 (\mathbf{g}_{\mathbf{k}_0} \mathbf{d}_{32})^2}{2\hbar^4} \theta^{(-)^2}(\omega_{\mathbf{k}_0}) \operatorname{sh} r \operatorname{ch} r[2|\alpha|^2$$

$$\times \exp(2i\varphi_{\alpha}) - \operatorname{sh} r \operatorname{ch} r \exp(i\varphi_{r})]L(\delta), \qquad (28)$$

$$L(\delta) = \begin{cases} \frac{1}{4K} \left| \frac{\sin(\delta/2K)}{\delta/2K} \right|, & \Gamma \ll K, \\ \frac{1}{8\Gamma} \left/ \left[\left(\frac{\delta}{2\Gamma} \right)^2 + \frac{1}{16} \right], & \Gamma \gg K. \end{cases}$$
(29)

It follows from Eqs. (26) to (28) that in a strong EMF the contribution from the spontaneous decay processes (Figs. 1,1) and the scattering processes (Figs. 1,2; 1,5) is relatively small as compared to the contribution from the induced processes (Figs. 1,6-1,9). Since the coefficients $a_{4}(r,\alpha)$ and $b(r,\alpha)$, determining the intensity of the induced processes, contain expression (29), the magnitudes of the spectral widths Γ and K have a large effect on the kinetics of the system in the case of an interaction with a strong EMF.

4. LOSS COEFFICIENT OF THE PHOTONS OF A SQUEEZED **ELECTROMAGNETIC FIELD**

We consider the change in the average number of photons per unit time in a beam of squeezed light (\mathbf{k}_0 mode) due to the absorption of photons by the atomic system and their reemission into other EMF modes. We call the expression

$$\mu = dn(\mathbf{k}_{0})/dt, \qquad (30)$$

the loss coefficient; here $n(\mathbf{k})$ is the average number of photons in the mode k. In accordance with Eqs. (1) and (14) we write down the general expression

$$\frac{d}{dt} n(\mathbf{k}) = \sum_{\beta=1}^{2} \frac{\mathbf{g}_{\mathbf{k}} \mathbf{d}_{\beta\beta}}{\hbar} \langle [U_{\beta}^{\beta}(t) + U_{\beta}^{\beta}(t)] \\ \times \{a_{\mathbf{k}}^{+}(t) [1 + \delta_{\mathbf{k}\mathbf{k}_{0}}(\operatorname{ch} r - 1)] \\ - \delta_{\mathbf{k}\mathbf{k}_{0}} a_{\mathbf{k}}(t) \operatorname{sh} r \exp[-i(2\omega_{0}t - \varphi_{r})] + \alpha^{*}(t) \delta_{\mathbf{k}\mathbf{k}_{0}} \}^{\flat} + \operatorname{h.c.}$$
(31)

Using Eqs. (14) to eliminate the EMF operators and taking (22), (26), and (27) into consideration we get for $\mathbf{k} = \mathbf{k}_0$, after some transformations,

$$\frac{d}{dt}n(\mathbf{k}_{0}) = -2i\Omega(r,\alpha)\langle R^{-}\rangle$$

$$+ \sum_{\mathbf{k}_{1}} \frac{(\mathbf{g}_{\mathbf{k}_{1}}\mathbf{d}_{31})^{2}(\mathbf{g}_{\mathbf{k}_{0}}\mathbf{d}_{32})^{2}}{\hbar^{4}} \{ [\theta^{(-)2}(\omega_{\mathbf{k}_{1}})$$

$$\times (1+|\alpha|^{2}+\mathrm{sh}^{2}r)\varphi(\omega_{21}-\omega_{0}-\omega_{\mathbf{k}_{1}})$$

$$-\theta^{(+)2}(\omega_{\mathbf{k}_{0}})(|\alpha|^{2}+\mathrm{sh}^{2}r)\varphi(\omega_{21}+\omega_{0}-\omega_{\mathbf{k}_{1}})]$$

$$\times \langle R^{+}R^{-}\rangle + \delta_{\mathbf{k}_{1}\mathbf{k}_{0}}\theta^{(-)2}(\omega_{\mathbf{k}_{0}})$$

$$(|\alpha|^{2}+\mathrm{sh}^{2}r+2|\alpha|^{2}\mathrm{sh}^{2}r+\mathrm{sh}^{4}r)\langle R^{+}R^{-}\rangle$$

$$\times \varphi(\omega_{21}-\omega_{0}-\omega_{\mathbf{k}_{1}})$$

$$-\delta_{\mathbf{k}_{1}\mathbf{k}_{0}}\theta^{(-)2}(\omega_{\mathbf{k}_{0}})(2|\alpha|^{2}\mathrm{sh}^{2}r+\mathrm{sh}^{4}r)\langle R^{-}R^{+}\rangle$$

$$\times \varphi(\omega_{21}-\omega_{0}-\omega_{\mathbf{k}_{1}})\} + \mathrm{h.c.} \qquad (32)$$

and in the case $\mathbf{k} \neq \mathbf{k}_0$

d

$$\frac{d}{dt}n(\mathbf{k}) = \sum_{\mathbf{k}_{1}} \frac{(\mathbf{g}_{\mathbf{k}_{1}}\mathbf{d}_{s_{1}})^{2}(\mathbf{g}_{\mathbf{k}}\mathbf{d}_{s_{2}})^{2}}{\hbar^{4}} \left\{\theta^{(-)2}(\omega_{\mathbf{k}_{1}})\right.$$

$$\times \left[1 + \delta_{\mathbf{k}_{1}\mathbf{k}_{0}}(|\alpha|^{2} + \mathrm{sh}^{2}r)\right]$$

$$\times \varphi(\omega_{21} - \omega_{\mathbf{k}} - \omega_{\mathbf{k}_{1}}) + \delta_{\mathbf{k}_{1}\mathbf{k}_{0}}\theta^{(+)2}(\omega_{\mathbf{k}_{0}})(|\alpha|^{2} + \mathrm{sh}^{2}r)$$

$$\times \varphi(\omega_{21} + \omega_{0} - \omega_{\mathbf{k}}) \right\} \langle R^{+}R^{-} \rangle + \mathrm{h.c.} \quad (33)$$

We write Eq. (32) in a simpler form. To do this we consider the change in the total number (in all modes) of photons in the system. Summing Eq. (31) over all k and using Eq. (32) and (33) we find

$$\frac{d}{dt} \left(\sum_{\mathbf{k}} n(\mathbf{k}) + 2\langle R_z \rangle \right) = -4a_3(r, \alpha) \langle R^+ R^- \rangle, \qquad (34)$$

$$\frac{d}{dt} \langle R_z \rangle = i\Omega(r, \alpha) \langle R^- \rangle - a(r, \alpha) \langle R^+ R^- \rangle$$

$$+ b(r, \alpha) \langle R^- R^+ \rangle + \text{h.c.}, \qquad (35)$$

where Eq. (35) was obtained from the kinetic equation (21).

It is clear that because there are scattering processes at the frequency $\omega_{\mathbf{k}} = \omega_{21} + \omega_0$ the expression within brackets on the left-hand side of (34) is not an integral of the motion.

The nonconservation of the "number of particles" is a consequence of the fact that each atom in the system is, with a probability determined by the coefficients $b(r,\alpha)$ and $a_3(r,\alpha)$, free to go from the $|1\rangle$ state to the $|2\rangle$ state while absorbing two quanta from the external EMF and afterwards, in an anti-Stokes scattering process (Figs. 1,2; 1,3), again returning to the $|1\rangle$ state while absorbing one photon from the squeezed EMF and emitting spontaneously a new photon. As a result of such a sequence the state of the atomic subsystem is unchanged but the number of photons in the system is reduced by two quanta. It is necessary to emphasize that notwithstanding the nonconservation of the "number of particles" the total energy of the system is an integral of the motion:

$$\frac{d}{dt}\left(\sum_{\mathbf{k}}\hbar\omega_{\mathbf{k}}n(\mathbf{k})+\hbar\omega_{2i}\langle R_{z}\rangle\right)=0.$$

Splitting off from Eq. (34) the term $dn(\mathbf{k}_0)/dt$, using (33), and changing from a summation to an integration we finally get

$$\mu = -2 \frac{d}{dt} \langle R_z \rangle - 2[2a_1 + a_2(r, \alpha) + 3a_3(r, \alpha)] \langle R^+ R^- \rangle.$$
 (36)

It follows from Eqs. (36) and (26) that in the case of a two-photon interaction the absorption of squeezed light is compounded from two-photon absorption as such when two light quanta are absorbed and from single-photon absorption (scattering); both processes are here of second order and thus must be taken into account. We note also that thanks to the scattering processes the absorption of light occurs not only when the system is excited, but also when it relaxes.

It follows from Eqs. (36) and (35) that the fluctuations of the EMF strongly affect the absorption process. Indeed, since the term $d \langle R_z \rangle / dt$, contained in Eq. (36), depends on the two-quantum Rabi frequency $\Omega(r,\alpha)$, until the atomic subsystem goes over into an equilibrium state it is just $\Omega(r,\alpha)$ which will mainly determine the absorption process of the squeezed light. In accordance with the properties of $\Omega(r,\alpha)$ considered in the preceding section we can thus conclude that the stronger the photons are bunched in the applied EMF, the stronger they are absorbed and the absorption will be a maximum in the case of phase squeezing, $2\varphi_{\alpha} = \varphi_r + \pi$. As the bunching of the photons in the squeezed EMF weakens, the absorption decreases and it reaches a minimum in the case of amplitude squeezing, $2\varphi_{\alpha}=\varphi_{r}$, and in the case when $\sinh r \cosh r=|\alpha|^{2}$ the fluctuations of the EMF strength become commensurable with the average value of the EMF strength and then the coefficient μ vanishes in first order. In the case of amplitude squeezing the quantum fluctuations of the squeezed EMF thus prevent its absorption whereas, in contrast, in the case of phase squeezing the quantum fluctuations accelerate the absorption of the EMF.

Let us assume that the atomic system is in a stationary state: $d \langle R_z \rangle / dt = 0$. To obtain quantitative results we must find the stationary value of the $\langle R^+R^- \rangle$ correlator. Following Ref. 14 we use a canonical transformation to change to new operators:

$$R^{-} = \Delta_2 S^{-} - \Delta_1 S^{+} + 2 (\Delta_1 \Delta_2)^{\eta_2} S_z,$$

$$S^{+} = (S^{-})^{+}, \quad 2S_z = [S^{+}, S^{-}],$$
(37)

 $\Delta_1 = \sin^2 \eta, \quad \Delta_2 = \cos^2 \eta, \quad \operatorname{ctg} 2\eta = 2\delta/G, \quad G^2 = \delta^2 + \Omega^2(r, \alpha).$

We assume that the external EMF is sufficiently strong:

$$G \gg Na(r, \alpha),$$
 (38)

i.e., we shall consider values of N and $a(r,\alpha)$ for which the two-quantum dynamical Stark effect occurs. Equation (23) then takes the form

$$\frac{dW_{A}}{dt} - iG[W_{A}, S_{z}] = \tilde{X}_{1}[S_{z}W_{A}, S_{z}] + \tilde{X}_{2}[S^{-}, W_{A}S^{+}] - \tilde{X}_{3}[S^{-}, S^{+}W_{A}] + \text{h.c.},$$
(39)

where

$$\widetilde{X}_1 = \Delta_1 \Delta_2 [a(r, \alpha) + b(r, \alpha) - d(r, \alpha) - d^*(r, \alpha)], \qquad (40a)$$

$$\widetilde{X}_{2} = \Delta_{2}^{2} a(r, \alpha) + \Delta_{1}^{2} b(r, \alpha) + \Delta_{1} \Delta_{2} [d(r, \alpha) + d^{*}(r, \alpha)]. \quad (40b)$$

$$\widetilde{X}_{3} = \Delta_{1}^{2} a(r, \alpha) + \Delta_{2}^{2} b(r, \alpha) + \Delta_{1} \Delta_{2} [d(r, \alpha) + d^{*}(r, \alpha)], \quad (40c)$$

and W_A is the statistical operator of the atomic subsystem after the transformation (37). We write the normalized stationary solution $(dW_A/dt = 0)$ of Eq. (39) in the form¹⁴

$$W_{A^{ss}} = Z^{-1} \sum_{n=0}^{N} X^{n} | n \rangle \langle n |,$$

$$X = \frac{\bar{X}_{2}}{\bar{X}_{3}}, \quad Z = \frac{X^{N+1} - 1}{X - 1}.$$

Taking into account what we have just said, Eq. (36) takes the form

$$\mu^{ss} = -2[2a_{1} + a_{2}(r, \alpha) + 3a_{3}(r, \alpha)] \times [(\Delta_{1}^{2} + \Delta_{2}^{2})\langle S^{+}S^{-}\rangle - 2\Delta_{1}^{2}\langle S_{z}\rangle + 4\Delta_{1}\Delta_{2}\langle S_{z}^{2}\rangle], \qquad (41)$$

$$\langle S^{+}S^{-}\rangle = N\left(\langle S_{z}\rangle + \frac{N}{2}\right) - - Z^{-1}\left[\frac{N(N+1)X^{N+1}}{X-1} - \frac{2(N+1)X^{N+2}}{(X-1)^{2}} + \frac{2(X^{2} - X^{N+3})}{(1-X)^{3}}\right], \qquad (42a)$$

$$\langle S_{z}\rangle = \frac{(N+1)X^{N+1}}{X^{N+1}-1} + \frac{X}{1-X} - \frac{N}{2}, \qquad (42a)$$

$$\langle S_{z}^{2}\rangle = \frac{N}{2}\left(\frac{N}{2} + 1\right) + \langle S_{z}\rangle - \langle S^{+}S^{-}\rangle. \qquad (42b)$$

Although μ^{SS} does not contain the phase-sensitive coefficients $\Omega(r,\alpha)$, $d(r,\alpha)$, and $d^*(r,\alpha)$, it depends implicitly (through the averages $\langle S^+S^- \rangle$, $\langle S_z \rangle$, and $\langle S_z^2 \rangle$) on the method of squeezing. One can show by using Eqs. (40), (41), and (42) that the rate of absorption of photons is larger for phase than amplitude squeezing.⁹

We obtained Eq. (41) for a high-intensity EMF; if, however, we consider an EMF of arbitrary intensity there is the possibility, without having recourse to condition (38), to study the r = 0 case, i.e., two-quantum absorption processes of a coherent EMF. Following Refs. 15 and 16 we find the stationary solution of Eq. (24):

$$\rho_{A^{**}} = B^{-1} \sum_{m,n=0}^{N} (g^{*})^{-m} (g)^{-n} a_{m,n} (R^{-})^{m} (R^{+})^{n},$$

$$B = \sum_{m=0}^{N} a_{m,m} H_{N,m} |g|^{-2m},$$

$$H_{N,m} = \frac{(N+m+1)! (m!)^{2}}{(N-m)! (2m+1)!},$$

$$a_{m,n} = \frac{\Gamma(m-\Delta+1)\Gamma(n+\Delta+1)}{m!n!\Gamma(1+\Delta)\Gamma(1-\Delta)},$$

$$g = \frac{i\Omega(\alpha)}{a(0,\alpha)}, \quad \Delta = \frac{i\delta}{a(0,\alpha)},$$

where $\Gamma(x)$ is the Euler gamma function and B a normalizing factor. The loss coefficient can then be written in the form

$$\mu^{**} = -2[2a_1 + a_2(0, \alpha) + 3a_3(0, \alpha)] \langle R^+ R^- \rangle,$$

$$\langle R^+ R^- \rangle = B^{-1} \sum_{\max(k, l)} (g^*)^{k-n} (g)^{l-n} a_{n-k,n-l} H_{N,n}.$$
(43)

Following Refs. 15 and 16 one can show that when there is no mismatch from resonance $(\delta = 0)$ the $\langle R_z \rangle$ and $\langle R^+R^-\rangle$ correlators, considered as functions of the parameter $\theta = \beta(\alpha)/N$ with $\beta(\alpha) = 2|\Omega(\alpha)|/a(0,\alpha)$, have singularities in the point $\theta = 1$; thus, the derivative of $\langle R_z \rangle$ in the point $\theta = 1$ tends to infinity while $\langle R^+ R^- \rangle$ has in the point $\theta = 1$ no uniquely defined derivative at all. This is the basis of speaking about a phase transition consisting in the transition of the atomic system from the usual state $\theta < 1$ to a state $\theta > 1$ where the two-quantum Stark effect occurs. However, since $a(0,\alpha)$, in contrast to the single-photon case,^{15,16} depends on the intensity of the applied EMF the behavior of the system has its own peculiarities. It follows from Eqs. (26) that the coefficient $a(0,\alpha)$ is the sum of three terms: a_1 , $a_2(0,\alpha)$, and $a_3(0,\alpha)$ describing the transition from the $|2\rangle$ state to the $|1\rangle$ state as the result of a twoquantum spontaneous decay and scattering of the applied EMF while in a strong EMF the contribution from the scattering processes is the decisive one. Neglecting the contribution from the spontaneous decay processes and noting [see (13) and (26)] that $a_2(0,\alpha)$, $a_3(0,\alpha)$, and the two-quantum Rabi frequency $\Omega(\alpha)$ are proportional to the intensity of the external EMF we can then conclude that the coefficient $\beta(\alpha)$ tends to saturation when the intensity of the coherent EMF increases:

$$\beta^{\infty} = \frac{3c^{3}\hbar}{2d_{31}d_{32}} \frac{(\omega_{31}-\omega_{0})(\omega_{32}+\omega_{0})}{(\omega_{31}+\omega_{32})(\omega_{21}-\omega_{0})^{3}} \times \left[1 + \left(\frac{\omega_{21}+\omega_{0}}{\omega_{21}-\omega_{0}}\right)^{3} \left(\frac{\omega_{31}-\omega_{0}}{\omega_{31}+\omega_{0}}\right)^{2} \left(\frac{\omega_{32}+\omega_{0}}{\omega_{32}-\omega_{0}}\right)^{2}\right]^{-1}.$$
(44)

It follows from what was said above that one of the conditions for the existence of a critical phase transition point, $\theta = 1$, in the case of two-photon interactions with strong coherent EMF takes the form $\beta^{\infty}/N = 1$, whence it follows that since β^{∞} is independent of the intensity of the applied EMF the correlators $\langle R_z \rangle$ and $\langle R^+R^- \rangle$ as functions of $\theta = \beta^{\infty}/N$ are, in fact, functions of the number of atoms in the system. Hence it follows, in turn, that there exists a critical value of the number of atoms $N_{\rm cr} = \beta^{\infty}$ and a phase transition is possible only when the number of atoms in the system is less than $N_{\rm cr}$ (Fig. 2b). In the opposite case, $N > N_{\rm cr}$, there cannot occur a phase transition, since we have always $\theta < 1$ (Fig. 2a).

Turning to Eq. (43) and taking into account the properties of $\langle R^+R^- \rangle$, shown in Fig. 2, we can conclude that the rate of photon absorption is a maximum in the vicinity of the point $\theta = 1$, i.e., when the number of atoms in the system is roughly equal to $N_{\rm cr}$. Outside the neighborhood of $\theta = 1$ $(N = N_{\rm cr})$ the rate of photon absorption is larger in the region where there is a two-photon Stark splitting, $\theta > 1$ $(N < N_{\rm cr})$ (Fig. 2b) than in the region where there is no Stark splitting, $\theta < 1$ $(N > N_{\rm cr})$ (Fig. 2a). It has thus been shown that in the case of interactions with a high intensity coherent EMF, when the above considerations are valid, the rate of light absorption μ^{ss} and the occurrence of a two-quantum dynamic Stark effect depend to a larger extent on the number of atoms in the system than on small changes in the intensity of the applied EMF.

In conclusion we note that although in the case of interactions with a high intensity EMF the main contribution



FIG. 2. The stationary values of the $\langle R^+R^-\rangle/(N/2)^2$ and $\langle R_z\rangle/N$ correlators as functions of the parameter $\theta = \beta^{\infty}/N$. We have taken $N_{\rm cr} = 2200$ as the critical value of the number of atoms in the system, obtained using (44) for the two-photon transition $2^2S_{1/2} \rightarrow 1^2S_{1/2}$ in Ar⁺¹⁷: a—N \geqslant 3200; b—N \geqslant 1000. The curves were drawn for a value $\delta = 0$.

comes from induced processes (Figs. 1,6; 1,9), it follows from Eq. (26c) that in the case where the $|3\rangle$ and the $|2\rangle$ levels are close to one another, $\omega_{32} - \omega_0 < \omega_0$, the contribution from scattering processes at the frequency $\omega_{\mathbf{k}} = \omega_{21} + \omega_0$ (Figs. 1,2; 1,3) becomes important. Since the collective scattering at the frequency $\omega_{\mathbf{k}} = \omega_{21} + \omega_0$ destroys the Stark splitting of the levels, in the case where the $|3\rangle$ level lies close to the $|2\rangle$ level, no two-photon dynamic Stark effect will not occur even in the $\theta > 1$ region.

5. CONCLUSION

The main aim of the present paper consisted in a study of two-quantum absorption processes of squeezed light. The results obtained supplement substantially the results of Ref. 9; the analysis of the two-quantum processes given in Ref. 9 is based on the approach proposed by Mollow in Ref. 17, but such an approach does not take into account scattering processes of the external EMF or the dynamics of the occupation of the levels of the atomic system, including collective effects in the system.

For an experimental observation of two-photon absorption of squeezed light one can use hydrogenlike or heliumlike atoms reaching two-photon resonance with the external EMF. For instance, one could use a system of Ar^{+17} atoms with the dipole-forbidden $2^2S_{1/2} \rightarrow 1^2S_{1/2}$ transition. The two-quantum spontaneous decay process was studied in Ref. 18 using such a system. We note that the linear dimensions of the atomic system must be smaller than the wavelength of the radiation of the applied EMF, since the space-time synchronism between reemitted photons plays a large role in extended systems.

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